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Vahid Salari, Christian Brouder,

Institut de Minéralogie et de Physique des Milieux Condensés, Université Pierre et Marie Curie-Paris 6, CNRS UMR7590, Boîte courrier 115, 4 place Jussieu, 75252 Paris cedex 05, France.

Abstract

Popp and Yan [F. A. Popp, Y. Yan, Phys. Lett. A 293 (2002) 93] proposed a model for delayed luminescence based on a single time-dependent coherent state. We show that the general solution of their model corresponds to a luminescence that is a linear function of time. Therefore, their model is not compatible with experimental delayed luminescence. Moreover, the functions that they use to describe the oscillatory behaviour of delayed luminescence are not solutions of the coupling equations to be solved.

Key words: Delayed luminescence; Hyperbolic decay; Coherent states; Biophotonics
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1. Introduction

Delayed luminescence is the phenomenon in which light-irradiated plants reemit photons during a long period of time, up to several minutes after the end of irradiation [1]. Delayed luminescence exhibits intriguing characteristics, such as hyperbolic (instead of exponential) decay and sometimes oscillations known as afterglow [1].

Two kinds of interpretation of delayed luminescence can be found in the literature (see Refs.[1–5] for recent references). The first one, which is usually not quantitative, uses the details of the photosynthetic process, in particular the dynamics of Photosystem II reaction centers. The second one, proposed by Popp and Yan [6], is based on the idea that a coherent state of the radiation field is present in living cells. The model seems to reproduce the hyperbolic decay of delayed luminescence as well as its oscillations.

In this comment, we extend the Popp and Yan model by diagonalizing the most general homeostatic time-dependent Hamiltonian compatible with single-mode coherent states. We show that this model cannot agree with experiment. In other words, the apparent agreement of the model with the hyperbolic decay of delayed luminescence is due to a calculation error in ref. [6].

Popp and Yan also consider hyperbolic decay to be the solution of a classical equation that oscillates with a constant frequency [7]. We show that a more general solution exists with a different asymptotic decay and that the hyperbolic solution is unstable. Moreover, we show that the oscillatory behaviour of delayed luminescence is not described by their model.

Finally, we stress the consequences of our findings for the modelling of delayed luminescence.

Email address: vahid.salari@impmc.upmc.fr (Vahid Salari).

2. Hyperbolic relaxation

The most general time-dependent Hamiltonian $H(t)$ which is solved by a single-mode (time-independent) coherent state $|v\rangle$ is [8]

$$H(t) = \omega(t)a^\dagger(t)a(t) + f^*(t)a(t) + f(t)a^\dagger(t) + \beta(t),$$

where $a(t)$ is the annihilation operator (in the Heisenberg picture), ω and β are real functions of time and f a complex function of time. As a coherent state, $|v\rangle$ satisfies $a(t)|v\rangle = \alpha(t)|v\rangle$ for some complex function α. The general form of $\alpha(t)$ is given by Mehta and Sudarshan [8] and reproduced (with two misprints) by Popp and Yan:

$$\alpha(t) = \mathrm{e}^{-i\psi(t)}\Big(\alpha(0) - i\int_0^t f(t')\mathrm{e}^{i\psi(t')}dt'\Big), \qquad (1)$$

where $\psi(t) = \int_0^t \omega(t')dt'$.

The mean number of photons as a function of time is given by $n(t) = \langle v|a^+(t)a(t)|v\rangle = |\alpha(t)|^2$. Thus,

$$n(t) = \Big|\alpha(0) - i\int_0^t f(t')\mathrm{e}^{i\psi(t')}dt'\Big|^2. \qquad (2)$$

By assuming "homeostasis" and the fact that "α should not be influenced by external classical energy sources" (we do not comment here on the validity of these assumptions), Popp and Yan obtain a condition on f described by the equation: $\dot f + i\omega f = 0$, with general solution $f(t) = f(0)\mathrm{e}^{-i\psi(t)}$.

By introducing this value of $f(t)$ into eq. (2), we obtain

$$n(t) = |\alpha(0) - if(0)t|^2. \qquad (3)$$

Therefore, $n(t)$ is a quadratic function of t, independent of $\omega(t)$.

The relation between the intensity of light $I(t)$ and $n(t)$ is not given explicitly in Ref. [6], but the caption of Fig. 3 shows that the authors use the relation $I(t) \propto \dot n(t)$. It follows from eq. (3) that $I(t)$ is a linear function of time. This does not agree with any measurement of delayed luminescence.

Therefore, the experimental hyperbolic relaxation is not accounted for by the coherent-state model of the paper.

The error in the paper by Popp and Yan is readily identified. The homeostasis hypothesis implies also the second equation $\omega\dot n + \dot\omega n + \dot\beta = 0$. Popp and Yan then choose $\omega_h(t) = \lambda/(1+\lambda t)$ and $\dot\beta$ to be time independent and they get a hyperbolic decay (up to an additive constant). The problem is that the second equation is in fact an equation for β (with obvious solution $\beta(t) = \beta(0) + \omega(0)n(0) - \omega(t)n(t)$), because n is already determined and ω is given. Note also that we worked with a general ω and not with the hyperbolic ω_h used by Popp and Yan.

It might be interesting to discuss the origin of this particular ω_h.

3. Origin of hyperbolic relaxation

Popp and Li obtained the hyperbolic solution ω_h as follows [7]. They start from the differential equation $\ddot x(t) + 2\mu(t)\dot x(t) + \omega_0^2 x(t) = 0$ and they use the classical method to remove the term in $\dot x$ by writing $x(t) = \exp\big(-\int_0^t \mu(\tau)d\tau\big)y(t)$. The equation for y is $\ddot y + (\omega_0^2 - \mu^2 - \dot\mu)y = 0$. They argue that the oscillating part $y(t)$ should have a constant frequency. For this, they solve $\mu^2 + \dot\mu = 0$ and get $\mu(t) = \lambda/(1+\lambda t)$ and

$$x(t) = y(t)/(1+\lambda t) = \omega_h(t)\frac{y(t)}{\lambda}.$$

However, even if we accept the requirement that the oscillating part has a constant frequency, then the equation that we have to solve is $\mu^2 + \dot\mu = \omega^2$, so that y oscillates with frequency $\sqrt{\omega_0^2 - \omega^2}$. The equation for μ has the solution $\mu(t) = \omega\tanh(\omega t + \mu_0)$ and $x(t) = y(t)\cosh\mu_0/\cosh(\mu_0 + \omega t)$. Of course, we can also consider $\mu^2 + \dot\mu = -\omega^2$, for which $\mu(t) = -\omega\tan(\omega t + \mu_0)$ and $x(t) = y(t)\cos\mu_0/\cos(\mu_0 + \omega t)$. The important point is that, in both cases, the decay is not hyperbolic and the limit of these solutions for $\omega \to 0$ is $\mu(t) = 0$. In other words, the hyperbolic solution is unstable and the slightest perturbation transforms it into a solution with a completely different asymptotic behaviour.

4. Oscillations

Popp and Yan claim that the oscillatory behavior of delayed luminescence can be explained by a coupling of two coherent states $|\alpha_1\rangle$ and $|\alpha_2\rangle$ described by differential equation (18'), where the primed equation numbers correspond to the equation numbers of ref. [6]. We shall see that this interpretation meets a rather serious inconsistency. In their calculation, they use eq. (18') to derive eq. (21') and solve eq. (21') with α_1 and α_2 defined by eq. (22'). However, these α_1 and α_2 are not solutions of the starting eq. (18'). In other words, their "solutions" do not solve the coupling equation. Their error comes from the fact that a solution of eq. (21') is generally not a solution of eq. (18'). Indeed, take any differentiable real function $y_1(t)$ and define $y_2 = y + y_1$, with

$$y(t) = \kappa \ln(1 + \lambda_1 t) - \kappa \ln(1 + \lambda_2 t) + \phi.$$

Then, $\alpha_1(t) = |a_1| e^{-iy_1(t)}$ and $\alpha_2(t) = |a_2| e^{-iy_2(t)}$ define a solution of eq. (21') which is (generally) not a solution of eq. (18').

5. Conclusion

We showed that the coherent state model proposed by Popp and Yan does not agree with delayed luminescence experiments, even if we generalize their approach: the math is simply wrong.

By fitting the reaction rates of the standard photosynthetic model, it was possible to quantitatively reproduce the short-time decay of plant luminescence (see [9] and references therein). However, no calculation of that kind was reported in the literature for long-time delayed luminescence, probably because of the apparent success of the model proposed by Popp and Yan. We hope that the present paper will clear the way for such studies.

To conclude with a positive remark, we want to stress that, even if the oversimplified model of a single coherent state must be dismissed, growing evidence shows that quantum coherence plays a role in photosynthesis [10,11]. Its influence on delayed luminescence remains to be investigated.

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